

## Determination of natural radioactivity concentration in consumed nuts and seeds and their Implications in the human body

Amal Hussain Alghamdi

King AbdulAziz University, Saudi Arabia

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The concentrations of natural radioactivity of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  were measured in nuts and seeds samples consumed in Saudi Arabia. A high-resolution HPGe detector was used for the natural radionuclides measurement. The results indicate that  $^{40}\text{K}$  was detected in all samples, whereas  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  were found only in Brazil nut sample. The average concentration of  $^{40}\text{K}$  in the investigated samples was  $363.82 \text{ Bq kg}^{-1}$ . The estimated annual effective dose due to ingestion of nuts and seeds was  $0.068 \text{ mSv y}^{-1}$  lower than the annual dose limit of  $1 \text{ mSv y}^{-1}$  for public exposure. This indicates that no risk is expected by the intake of the studied nuts and seeds samples. The radionuclide concentrations were compared with those reported from different countries. A study was carried out to evaluate the association of levels of radioactivity, selenium and aflatoxin in shelled Brazil nuts, which were classified in different sizes, for export. The selenium determinations were performed by inductively coupled plasma optical emission spectrometry (LOQ =  $3.0 \mu\text{g g}^{-1}$ ), and aflatoxins were detected by Liquid chromatography-mass spectrometry (LOQ =  $0.85 \mu\text{g kg}^{-1}$ ), recovery rates were between 92 and 100%. Radioactivity was measured by high-resolution gamma spectrometry. The selenium mean concentration was ( $22.7 \pm 7.4$ )  $\mu\text{g g}^{-1}$ . ( $n = 30$ ). Mean activities determined for the following radium isotopes were:  $15.77 \text{ Bq kg}^{-1}$  for  $^{224}\text{Ra}$ ,  $104.8 \text{ Bq kg}^{-1}$  for  $^{226}\text{Ra}$  and  $99.48 \text{ Bq kg}^{-1}$  for  $^{228}\text{Ra}$ . For  $^{226}\text{Ra}$ , the levels did not vary significantly with nut sizes, although such differences were observed for  $^{224}\text{Ra}$  and  $^{228}\text{Ra}$ . There was no statistically significant association between the level of selenium and the activity of radionuclides, however, there was correlation between the radionuclides. Aflatoxins above the quantification limit were not

found. The concentration of natural radioactivity in rice is an important parameter for the determination of population exposure by the ingestion of natural radionuclides during habitual consumption of food. All types of food including rice contain a detectable amount of natural radioactivity which successively relocate into the human body via the ingestion pathway. Rice is the main cultivated crop in Bangladesh and most of the Bangladeshi people consume rice as their staple food. Hence, studies on the evaluation of natural radioactivity in rice have been performed by gamma-ray spectrometry using High Purity Germanium (HPGe) detector in order to estimate various radiation hazards due to rice consumption.

The average activity levels of natural radionuclides  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$  in the rice samples were  $1.09 \pm 0.31$ ,  $0.17 \pm 0.21$  and  $4.70 \pm 1.59 \text{ Bq kg}^{-1}$ , respectively. The estimated effective doses for the respective radionuclides caused by the rice consumption were  $43.69$ ,  $16.39$  and  $4.15 \mu\text{Sv y}^{-1}$ , respectively which was below the UNSCEAR compiled value. The calculated excess lifetime cancer risk (ELCR) values via rice consumption were found below the acceptable limit of  $0.29 \times 10^{-3}$  for radiological risk. The analysis of selenium was performed by ICP optical emission spectrometry (OES), using the atomic emission (28). The digestion of samples was carried out by acid in microwave in a closed system. Mass of about  $0.3 \text{ g} + 3 \text{ mL}$  of concentrated nitric acid and  $1 \text{ mL}$  of hydrogen peroxide, high pressure system 100 DAK Bergof, irradiation with 200 watts for five minutes and increase 50 watts  $\text{min}^{-1}$  up to 700 watts for fifteen minutes, total time thirty minutes. It was then cooled, depressurized and swelled to  $50 \text{ mL}$ . The limit of detection (LOD) was  $1.50 \text{ mg g}^{-1}$  and the limit of quantification (LOQ) was  $3.00 \text{ mg g}^{-1}$ . The LOQ was defined as the lowest point of the curve with high reproducibility,

axial view. The level of recovery was 90% (n=30). The analytical lines used for selenium determination was 196.03 nm. The operational features of the spectrometer were: operating range optics - Used: 170 to 850 nm, power radio frequency = 1300 watts box, plasma gas flow rate = 15 L min<sup>-1</sup> - adjunct = 2 L min<sup>-1</sup> - 0.6 nebulizer mL min<sup>-1</sup>, peristaltic pump with adjustable flow rate of 2 mL min<sup>-1</sup>. flatoxin determination was performed by liquid chromatography (LC) coupled with tandem - mass spectrometry (MS-MS) APCI in the positive detection mode (Xavier and Scussel 2008). The LC conditions (C8 column) involved a mobile-phase with a methanol/water gradient [45% water/55% methanol (three minutes); from three to five minutes the gradient was changed to 30% water/70% methanol] and a flow rate of 1 mL min<sup>-1</sup>. For MS/MS, the parent and two daughter ions (m/z) were selected for each toxin as follows: AFB1, m/z 313.1 (241.10 and 285.10); AFB2, m/z 315 (259.09 and 287.20); AFG1, m/z 329.1 (200.05 and 243.05); and AFG2, m/z 331.2 (245.07 and 231.20). The LOD and LOQ values for LC-MS/MS of AFB1, AFB2, AFG1, AFG2 were 0.05, 0.075, 0.075 and 0.1 µg kg<sup>-1</sup> and 0.15, 0.2, 0.2 and 0.3 µg kg<sup>-1</sup> for each aflatoxin, respectively. The LOD and LOQ values for total aflatoxin were 0.3 and 0.85 µg kg<sup>-1</sup>. To obtain those parameters, the finely ground Brazil nuts were homogenized and spiked prior to extraction with aflatoxins at five concentrations between 1 to 10 µg kg<sup>-1</sup>. Portions of

25 g were taken for extraction by adding 100 mL of acetonitrile/water (80:20, v/v) to the sample, which was followed by mixing for 2 h and filtration. The LOD method was defined by t times the signal-to noise ratio, and the LOQ method was defined by 6 times the signal-to-noise ratio. Five-point analytical curves were constructed for quantification and for the estimation of LOD and LOQ. Each point corresponded to a mean of five injections of each extract. The recoveries for each aflatoxin (AFB1, AFB2, AFG1, AFG2) were 92.4, 72.5, 99.8, and 97.1%, respectively.

The shell/nut ratio used for calculation was that reported by de Mello and Scussel [60:40 (60% shell/40% nut) with a factor of 1.5, which was considered the standard ratio for normal healthy Traces of the radionuclides 228Ra, 226Ra and 224Ra were measured by gamma spectrometry using a HPGe detector (GEM-M 7080-PS, ORTEC) with 66% relative efficiency. Each nut sample was counted for 86,400 seconds. The concentration of 228Ra was determined from the 911, 338 and 969 keV lines for 228Ac in each sample. The concentration of 226Ra was determined from the 609, 1120 and 1764.5 keV lines for 214Bi, and from the 352 and 295 keV lines for 214Pb. The average activity of each of these two radionuclides in all samples was then calculated. Subsequently, an average weighted by the variances of these two values was used to determine the average value of the activity of 226Ra and 228Ra in these nuts.